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U.S. GEOLOGICAL SURVEY
WATER RESOURCES DIVISION
NATIONAL RESEARCH PROGRAM
PROPOSAL TO DEPARTMENT OF ENERGY

A. TITLE: Determination of colloidal and solution-phase transport of americium and plutonium in groundwater at Rocky Flats

B. PROBLEM: Colloidal particles have been observed in groundwaters (Degueldre et al., 1989; Salbu et al., 1985). Colloidal-size particles have been shown to be mobile in aquifers (Harvey et al., 1989). Association of insoluble contaminants with groundwater colloids is expected to greatly influence contaminant transport (McCarthy and Zachara, 1989; Mills et al., 1991). A number of studies has shown the importance of this process in radionuclide transport (Buddemeier and Hunt, 1988; Penrose et al., 1990).

The presence of colloidally-associated americium and plutonium must be assessed in order to predict both the groundwater transport and the potential effectiveness of remediation plans. The difficulty in determining the chemical nature of groundwater colloids is a result of their low concentrations. Cross-flow filtration has been applied to the collection of large quantities of colloids for characterization studies (Rees and Ranville, 1990, Ranville et al., 1991). A more complete understanding of the size distribution and the chemical phases of americium and plutonium-bearing colloids will improve predictive capabilities for both groundwater transport (Harvey and Garbedian, 1991; Hunt et al., 1987; Travis and Nuttal, 1987) and the effectiveness of remediation procedures such as filtration (Tobiason and O'Melia, 1988).

C. RESEARCH OBJECTIVES: The research objectives of this investigation are to determine the dominant phases controlling the transport of contaminants in groundwater at the Rocky Flats site. Specific research objectives include: (1) determination of chemical characteristics and mineralogy of colloidal material in surface water and groundwater samples, as related to the affinity of colloidal material for sorption of americium and plutonium; (2) determination of distribution of americium and plutonium between solution and colloidal phases of varying sizes; (3) determination of the variation along a groundwater flow path in the distribution of americium and plutonium between solution and colloidal phases; and (4) interpretation of the results of phase characterization in regards to the behavior of americium and plutonium during groundwater transport and remediation. Even if the concentrations of plutonium and americium are undetectable, this research will advance the overall understanding of colloids in groundwater and will be useful for planning future remediation activities at Rocky Flats.

D. APPROACH:

1. Tangential-flow filtration will allow processing of large volumes of groundwater in order to isolate colloids in sufficient quantity to perform extensive phase characterization. Sampling procedures

(i.e. pumping rates, redox conditions, etc.) will be used to insure that colloids are not generated during sampling. Use of different porosity filters will provide size distributions of colloids in the range of 0.005 to 1.0 micron. Eventual use of specially-fabricated track-etched membranes will provide much better size distribution data. Sequential extractions of colloids will provide information on the mineralogical and chemical phases and the extent of americium and plutonium association with these phases. SEM investigation of colloid concentrates will also provide chemical and mineralogical information.

2. The radiochemical analyses will be done by a DOE contractor (e.g. Los Alamos National Laboratory). The procedures for processing and analysis of the colloid and aqueous samples will be consistent with potential hazards as indicated by prior data from the site and the results of the radiochemical analyses. Chemical characterization of colloidal phases will include the following analyses: (1) element composition by various combustive techniques for C, H, O, N, S and P and by digestion and ICP analysis for major and trace inorganic constituents, (2) scanning electron microscopy (SEM) to determine aggregation and other morphological characteristics and EDX (energy dispersive X-ray analysis) for composition of mineral grains, (3) particle size and surface charge by light scattering and electrophoretic mobility, and (4) organic carbon and lipid content. Some of these analyses will be done by contract laboratories (depending on the nature of the samples, either Huffman Laboratories or Pace Laboratories, which is licensed to receive radioactive samples). If a relationship between Am and Pu content and organic carbon content is observed, GC/MS analysis of organic material in colloids will be performed in our lab or Pace Laboratories. The solution phases will be analysed for pH, major cations and anions, and dissolved organic carbon. Depending on the radiochemical results, oxidation state analyses of dissolved plutonium will also be done for the solution phase. In the final year of the study, a sequential extraction procedure will be developed and field flow fractionation will be used to refine size determinatins.

3. A more extensive field study will be designed based upon the initial results, and will involve collection of samples from an array of existing wells. Sampling will include wells along known hydrologic flowpaths.

4. Results will be modeled with MINTEQ (a chemical equilibrium computer program) to assess the importance of solubility controls, including polynuclear complexes, relative to sorption by colloids. The field study will be interpreted based upon existing information on hydrologic and mineralogical properties of the aquifer.

E. REPORTS: The information collected in this study will be maintained in USGS and DOE files, as described in the Interagency Agreement (IA), which is attached. Interpretive reports presenting results of the study will be prepared as stipulated in the IA. An initial USGS Open-File report will be prepared presenting the results of the initial characterization of the chemistry and mineralogy of colloidal material from the first phase of the study. A second report (potentially a journal article) from the second phase of the study will present the results from the distribution of americium and plutonium between colloidal and solution phases in groundwater and interpret these results based upon geochemical processes. A third report will present the results on the variation in the americium and plutonium distribution along a groundwater flow path.

F. BENEFITS: The investigation will utilize new analytical methods to quantify the role of colloidal transport of trace inorganic contaminants in groundwater and surface water systems. These results will be directly useful in management of environmental activities at the Rocky Flats facility, and will also contribute to the understanding of colloidal transport processes in other aquatic systems.

G. PERSONNEL and QUALIFICATIONS: Scientists working directly on the project will be Diane McKnight (project chief), Jim Ranville and Richard Harnish from the USGS, Water Resources Division, National Research Program (NRP). Laboratory assistance will be provided by Michael Anthony (USGS-NRP). This group has studied colloidal transport processes in diverse environments over the past several years including: (1) acid mine drainage streams, (2) Pueblo Reservoir, (3) Mississippi River, (4) wetlands, and (5) an Antarctic desert lake. Field sampling at the Rocky Flats site will be conducted in coordination with personnel designated by DOE and with other researchers working at the site. Initial results will be discussed with other scientists involved in particular aspects of the studies at Rocky Flats.

H. TIMELINE FOR COMPLETION OF STUDY: The objectives for Phase 1 will be carried out in the period August 1991 through April 1992. The objectives for Phase 2 will be carried out in the period May 1992 through March 1993.

Phase 1: Research Objectives

1. Purchase and assemble single-stage tangential-flow filtration system including environmental chamber.

2. Preliminary sampling of groundwater (2-3 sites) and isolation of colloids.

3. Submit colloid samples and filtrate for radiochemical and bulk chemical analysis.

4. Perform SEM examination of colloids.

5. Evaluate preliminary sampling procedure and results of colloid bulk chemical and radiochemical analysis, and prepare initial report.

6. Purchase, assemble and test sequential tangential-flow filtration system with track-etched membranes to refine procedures, based upon results from preliminary sampling.

Phase 2: Research Objectives

1. Execute more extensive sampling of a number of wells and seeps and isolate colloids in the spring and summer.

2. Submit colloid samples and filtrate for bulk chemical and radiochemical analysis.

3. Evaluate results from extensive sampling and chemical analysis.

4. Develop a sequential extraction procedure for phase determinations of colloids and associated americum and plutonium.

5. Perform sequential extractions of colloids and submit extractions for analysis.

6. Evaluate results in the context of remediation and groundwater transport, and prepare interpretive report.

I. FUNDING:

	Phase 1	Phase 2	
GS 15	1 mo. $5,332	1 mo. $5,332	
GS 12	6 mo. $22,308	12 mo. $48,334	
GS 7	9 ~~14~~ mo. ~~$31,440~~ 20,211	12 mo. $27,248	
Equipment	50,000	--	
Supplies	7,000	7,000	
Analyses*	15,000	30,000	
Travel	7,000	7,000	
NRP overhead (40%)	~~55,232~~ 50,740	49,966	
TOTAL:	~~193,312~~ 177,591	174,880 →	$352,471.00

* not including radiochemical analyses

J. REFERENCES:

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Same as Hunt?